

The influence of extremely large solar proton events in a changing stratosphere

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Short title: STRATOSPHERIC INFLUENCE OF SOLAR PROTON EVENTS

Abstract. Two periods of extremely large solar proton events (SPEs) occurred in thirty years, which forced significant long-term polar stratospheric changes. The 1957-10, 1972 and October 19-27, 1989 SPEs happened in stratospheres that were different chemically. The stratospheric chlorine levels were relatively small in 1972 (~ 1 ppbv) and were fairly substantial in 1989 at about (~ 3 ppbv). Although these events produced both HO_x and NO_y constituents in the mesosphere and stratosphere, NO_y constituents had lifetimes long enough to affect ozone for several months past the events. Our recently improved two-dimensional chemistry and transport atmospheric model was used to compute the effects of these gigantic SPEs in the stratosphere. Significant upper stratospheric ozone depletions $> 10\%$ are predicted to last for a few months past these SPEs. The long-lived SPE-produced NO_y constituents were transported to lower levels during winter after these huge SPEs and caused impacts in the middle and lower stratosphere. During periods of high halogen loading, these impacts resulted in interference with the chlorine and bromine loss cycles and ozone destruction. The chemical state of the atmosphere, including the stratospheric aerosol density, substantially affected the predicted stratospheric influence of extremely large SPEs.

Abstract

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Introduction

The stratosphere changed dramatically in the 1965-95 time period. Stratospheric chlorine levels rose by over a factor of three from about 1 ppbv in 1965 to around 3.3 ppbv in 1995 [WMO 1990, 1999]. Stratospheric sulfate aerosol density (SAD) amounts varied significantly in the 1979-95 time period, being at or near background levels during some years but increasing strikingly after major volcanic eruptions [e.g., Thomason *et al.*, 1997a,b]. Increases in either chlorine or SAD amounts can lead to halogen chemistry being more important in ozone control. These temporal stratospheric chemical changes can impact the effects of other processes, such as solar proton events.

Solar proton events (SPEs) are natural occurrences which can produce large amounts of HO_x (H, OH, HO_2) and NO_y (N, NO, NO_2 , NO_3 , N_2O_5 , HNO_3 , HO_2NO_2 , ClONO_2 , BrONO_2) constituents in the middle atmosphere polar regions over very short periods of time (hours to days). The SPE-produced HO_x and NO_y constituents can lead to ozone decreases in the mesosphere and stratosphere [e.g., Swider and Keneshea 1973; Frederick 1976; Solomon *et al.*, 1983; Jackman and McPeters, 1985; Reid *et al.*, 1991; and Jackman *et al.*, 1995]. The HO_x constituents are relatively short-lived with lifetimes of only hours in the middle atmosphere, thus their atmospheric influence does not extend much beyond the duration of the SPE. However, the SPE-produced NO_y constituents can be transported downwards where their lifetimes range from months to years. These SPE-related NO_y constituents can then influence the middle and lower stratosphere over longer periods of time, but are also significantly affected by the levels of chlorine or

SAD amounts.

We recently studied the influence of SPEs in our two-dimensional (2-D) chemistry and transport atmospheric model in an assessment of the various components that were responsible for ozone change between 1975 and 1995 [*Jackman et al.*, 1996; abbreviated *J96*]. We found that the influence of SPEs on global total ozone in this time period was significant only after the extremely large October 1989 event. Both a decrease (for about 2 years after the event) and then an increase (from 3-5 years after the event) in global total ozone were predicted after this event. These simulated increases in ozone were caused by SPE-produced NO_y constituents being transported downwards and interfering with the chlorine and bromine loss cycles for ozone destruction. Our earlier study [*Jackman et al.*, 1995; abbreviated *J95*] did not uncover this process because: 1) the stratospheric sulfate aerosol density (SAD) used in *J95* was smaller than that employed in *J96*; 2) polar stratospheric clouds were not included in *J95* but were in *J96*; and 3) the transport formulation of our 2-D model was modified substantially between *J95* and *J96*.

The August 1972 SPE (not simulated in the modeling studies of *J96*) was also an extremely large event, which created significant NO_y to affect the stratosphere for months to years past the event [e.g., *Crutzen et al.*, 1975; *Heath et al.*, 1977; *Fabian et al.*, 1979; *Solomon and Crutzen*, 1981; *Reagan et al.*, 1981; *Rusch et al.*, 1981; *McPeters et al.*, 1981; *Jackman and McPeters*, 1987; *Jackman et al.*, 1990]. Although it was clear in earlier work on this gigantic SPE that the inclusion of chlorine chemistry was important in assessing the longer term impact of this event [first used in *Solomon and*

Crutzen, 1981], other previous studies, including ours [*Jackman et al.*, 1990; abbreviated *J90*], used only 'gas phase' chemistry in model simulations. Inclusion of heterogeneous chemistry processes on both the stratospheric SAD burden and polar stratospheric clouds are very important [see *WMO* 1992, 1995, 1999] in assessing influences on stratospheric ozone, thus the simulations given in *J90* might be substantially affected. The SPE-caused increases in ozone simulated in *J96* a couple of years after the October 1989 SPE should be sensitive to the stratospheric halogen loading and SAD burden, both of which affect the importance of halogens in influencing ozone.

Here we revisit some of this previous work [*J90*, *J95*, and *J96*] on the influence of SPEs on the stratosphere. We investigate the effects of SPEs over the 1963 to 1995 period with our improved 2-D model, focussing especially on the impact of the extremely large SPEs of August 1972 and October 1989 on the polar stratosphere. We extend the study of *J96* and investigate the sensitivity of these SPEs due to the halogen and SAD burden. These sensitivity computations allow us to quantify the importance of huge SPEs within the context of a changing stratosphere, both from mankind-related chlorine and bromine emissions and from volcanically-produced sulfate aerosols.

Proton Flux Data; HO_x and NO_y Production

We use solar particle fluxes from the Interplanetary Monitoring Platform (IMP) series of satellites over the time period 1963 through 1993. The solar proton fluxes for 1963 through 1973 were provided by T. Armstrong and colleagues (University of Kansas, private communication, 1986; also, see *Armstrong et al.*, 1983) for relatively coarse

intervals (> 10 MeV, > 30 MeV, > 60 MeV). These solar proton fluxes were used to compute daily average ion pair production profiles using the methodology discussed in *J90* for 1963 through 1973.

T. Armstrong and colleagues (University of Kansas, private communication, 1994) also provided the solar proton and alpha particle fluxes for 1974 through 1993, but for the very fine energy intervals detailed in the Appendix of *Vitt and Jackman* [1996; abbreviated *V96*]. These solar particle fluxes were used to compute daily average ion pair production profiles using the methodology discussed in *V96* for 1974 through 1993.

Although SPEs can occur at any time during the solar cycle, they tend to be more prevalent near solar maximum. The Earth's magnetic field affects solar protons and allows full access for these particles to the entire polar cap region (poleward of a cutoff geomagnetic latitude of about 60°) with relatively minor access outside this region. The polar cap region can change slightly depending on the spectrum of the protons. Higher energy protons tend to produce larger polar cap regions, *e.g.*, *Reagan et al.* [1981] noted that the August 1972 SPE caused a cutoff geomagnetic latitude of 58° . For the purposes of this study we assume that the solar protons deposit their energy in both hemispheres at geomagnetic latitudes above 60° . The geomagnetic poles are offset by about 11° from the geographic poles. Since our model is based on a geographic latitude coordinate, this results in weightings for particle influences of 1.0 for model grid boxes centered on $\pm 85^\circ$, 0.98 for $\pm 75^\circ$, 0.60 for $\pm 65^\circ$, 0.31 for $\pm 55^\circ$, 0.02 for $\pm 45^\circ$, and 0.0 for all other latitudes.

The precipitating particles primarily lose their energy in the creation of ion pairs in the atmosphere. Complicated ion chemistry following this ion pair production results in

the formation of HO_x constituents. Each ion pair produces about two HO_x constituents up to an altitude of approximately 70 km. Above 70 km, the production is less than two HO_x constituents per ion pair [Solomon *et al.*, 1981]. We use the production rates of HO_x constituents per ion pair as provided in [Solomon *et al.* [1981, Figure 2] in all our model computations [also, see Goldberg *et al.*, 1995]. The HO_x constituents, with lifetimes of only hours in the middle atmosphere, can deplete ozone in the mesosphere and upper stratosphere.

The precipitating particles and associated secondary electrons also produce atomic nitrogen via dissociations, predissociations, or dissociative ionizations in collisions with N₂. We assume that 1.25 N atoms are produced per ion pair for all our model computations [e.g., see Porter *et al.*, 1976 and Jackman *et al.*, 1980]. The N atoms rapidly produce other odd nitrogen (NO_y) constituents, which can then lead to ozone decreases in the middle and upper stratosphere. Since NO_y constituents have a relatively long lifetime of months to years in the middle and lower stratosphere, the transport of these species to lower levels is quite important in assessing their long-term influence on ozone. Our 2-D modeling studies will primarily focus on the impact of the SPE-produced NO_y on ozone.

We have quantified middle atmospheric NO_y production before [Jackman *et al.*, 1980; J90; and V96]. We show a summary of those computations of NO_y molecules produced per year for the northern polar stratosphere (latitudes > 50°N) in Figure 1 over the 1955 to 1993 time period. Several sources of NO_y are given in Figure 1 including SPEs, galactic cosmic rays (GCRs), ‘in situ’ oxidation of N₂O, and horizontal

transport from lower latitudes. The annual production rates of NO_y from SPEs for the periods 1955-1973 and 1974-1993 are taken from *J90* and *V96*, respectively. The full histograms indicate the SPE-caused NO_y production for both the stratosphere and mesosphere, while the light gray shaded areas of the histograms indicate the SPE-caused stratospheric NO_y production only.

The annual production rates of NO_y from GCRs, 'in situ' oxidation of N_2O , and horizontal transport from lower latitudes were all taken from *V96* and are represented in Figure 1 by the dash-dot-dot-dot, dash-dot, and dashed lines, respectively. *V96* compute these NO_y production rates for the 1974-93 time period and show only about a 10% variation in the 'in situ' oxidation of N_2O and horizontal transport from lower latitudes. This minor variation is hardly noticeable on the log scale used for the ordinate in Figure 1, and we assume constant NO_y production values of 1×10^{33} and 9×10^{33} NO_y molecules/year for the 'in situ' oxidation of N_2O and horizontal transport of NO_y from lower latitudes, respectively. The GCR contribution to NO_y was computed by *V96* to vary from 6.9 to 9.6×10^{32} molecules/year in the 1974-1993 time period. The GCR values shown in Figure 1 for the 1955-73 period were extrapolated assuming a similar temporal behavior.

The horizontal transport of NO_y from lower latitudes is due to the oxidation of N_2O and is about an order of magnitude larger than the NO_y sources of GCRs and 'in situ' oxidation of N_2O . The NO_y from SPEs is computed to vary by orders of magnitude over the 1955-1993 time period and there were only two years (1972 and 1989) when the annual SPE source of NO_y in the Northern Hemisphere was larger than 1.2×10^{33} NO_y

molecules/year. Huge fluxes of extremely energetic particles characterized the SPEs of August 1972 and October 1989 which dominated the NO_y sources for those years. *Zadorozhny et al.* [1992] measured huge NO enhancements of 2.6×10^{15} molecules/ cm^2 between 50 and 90 km at southern polar latitudes as a result of the SPE during the time period October 19-23, 1989. We compute an NO increase of 3.0×10^{15} molecules/ cm^2 over the same altitude and time range, thus there is reasonable agreement between measured and predicted NO_y production due to this gigantic SPE.

Two-dimensional Model Description and Primary Simulations

We used the latest version of the NASA Goddard Space Flight Center (GSFC) 2-D photochemical and transport model described in *J96*. The GSFC 2-D model was originally described by *Douglass et al.* [1989] and extended to mesospheric heights by *Jackman et al.* [1990]. Its vertical range, equally spaced in log pressure, is from the ground to approximately 90 km (0.0024 mbar) with approximately a 2-km grid spacing. Latitudes range from 85°S to 85°N with a 10° grid spacing. The model includes heterogeneous processes on the stratospheric sulfate aerosol layer and polar stratospheric clouds as well as 'gas phase' only chemical reactions.

Significant improvements have been made to the model transport formulation in the past few years [*J96*; *Fleming et al.*, 1999]. The climatological transport fields are based on empirical data sets. A 17-year average (1979-1995) of temperature data from the National Centers for Environmental Prediction (NCEP) was used to derive climatological temperature fields and planetary wave statistics used in the computation

of advection and horizontal diffusion fields. The transport changes daily, but repeats yearly. Further details of the transport formulation can be found in *J96* and *Fleming et al.* [1999].

We used the 2-D model to compute several time-dependent simulations over the time period 1960-1995. The two primary simulations were “Base” and “Perturbed,” shown in Table 1. The “Base” simulation did not include solar proton events, but did include 1) chlorine and bromine source gas boundary conditions from *WMO* [1990] for years prior to 1970 and from *WMO* [1999] for years 1970-95; 2) solar UV flux variations from *J96*; 3) stratospheric SAD variations from *J96*; and 4) a mean GCR flux from *J96*. The “Perturbed” simulation was the same as “Base” except it also included solar proton events.

Results and Discussion

Using “Base” and “Perturbed” simulation results, we computed the percentage change of constituents in the “Perturbed” simulation relative to the “Base” simulation. The major focus of this study is on the time periods after the extremely large SPEs of August 1972 and October 1989, thus we present the percentage total ozone change from our computations for the time periods 1972-75 and 1989-92 in Figures 2 and 3, respectively. The shaded areas indicate computed decreases in ozone, whereas the white areas indicate computed increases in ozone. Both the August 1972 and October 1989 SPEs caused decreases of over 1% in the southern and northern polar regions. Such small decreases would be very difficult to observe in total ozone measurements

given the large fluctuations observed over short periods of time (days to months) due to meteorological and seasonal variations [see *WMO* 1992, 1995, 1999]. *Marin and Lastovicka* [1998] were not able to find any signal for SPEs in a Dobson total ozone record for central Europe (50°N). There are some obvious differences in the response of total ozone to these huge SPEs in the two time periods with more areas of computed increases of ozone after the October 1989 SPE.

August 2-10, 1972 Solar Proton Event

A more detailed look at the predicted temporal behavior of profile O_3 and NO_y is given in Figure 4 for the time period 1972-75 at 75°N. We focus on 75°N as this latitude is primarily within the polar cap region affected by the solar protons. Predicted upper stratospheric increases in NO_y over 100% during and shortly after the August 1972 SPE cause upper stratospheric O_3 decreases of about 20%. The protons used in the computations of NO_y production for the August 1972 events had energies only up to 100 MeV [J90]. This artificial cutoff was required as the IMP proton flux measurements in 1972 provided only integrated flux data for the highest energy protons, those with energies > 60 MeV. We used an empirical formula to fit the proton flux data given and applied it over a 5-100 MeV range [see J90]. Although the 1972 SPE probably had protons with energies greater than 100 MeV, they could not be reliably included. Thus energy deposition of these particles and, subsequently NO_y production, did not occur below about 10 mbar.

Over time the large enhancements of NO_y in the upper stratosphere were

transported to lower levels causing ozone decreases in the lower stratosphere. Ozone decreases down to about 100 mbar were predicted by the spring of 1973 as a result of this event in August 1972. In a previous study [J90] we compared the short-term (August-September, 1972) predicted ozone change with Nimbus 4 Backscatter Ultraviolet (BUV) measurements and obtained fairly reasonable agreement (see Figures 6a and 7a of J90). Our predicted upper stratospheric ozone change for the time period up to two months past the August 1972 SPE has not changed significantly from that study, thus we do not repeat the comparison. Although we predict long-term stratospheric changes in NO_y and O_3 lasting for over three years past these large events, the NO_y increases are less than 5% and the O_3 decreases are less than 2% within six months of the SPEs.

Small increases in ozone ($\sim 2\%$) are predicted to occur near 10 mbar during and shortly after the gigantic August 1972 SPE (Figure 4). The increases in O_3 result from the well-known process of 'self-healing.' The 'self-healing' of ozone is caused when O_3 decreases above an atmospheric level permit increased ultraviolet light to lower levels that lead to O_2 dissociation and associated O_3 production below. Ozone 'self-healing' has been discussed before [see, *e.g.*, CIAP, 1975; Jackman and McPeters, 1985].

The temporal behavior of NO_y at 2 mbar, O_3 at three levels (2, 12, and 30 mbar), and column ozone at 75°N for 1972 is given in Figure 5. The very clear anti-correlation between NO_y increases and O_3 decreases is apparent at 2 mbar. At the lower levels, both increases and decreases in ozone are visible. Predicted ozone increases in August and September at 12 mbar are a result of 'self-healing' and predicted decreases for October through December are a result of the downward transport of ozone-destroying

NO_y to this lower level. The increases in ozone at 30 mbar resulting in a peak ozone change of +0.3% in November 1972 are a result of the region of decreased ozone above this level causing 'self-healing' at this even lower altitude (also, see Figure 4). Note, however, that the calculated changes at 30 mbar are much smaller than those at 12 mbar. The computed column ozone in the last five months of 1972 in the "Perturbed" simulation is always less than that in the "Base" simulation due to the August solar events (bottom plot of Figure 5). 'Self-healing' of ozone in the lower stratosphere only partially compensates for the higher altitude depletion of ozone caused by the SPE-generated NO_y .

October 19-27, 1989 Solar Proton Event

A more detailed view of the predicted temporal behavior of profile O_3 and NO_y for 1989-92 at 75°N is shown in Figure 6. Similar to the August 1972 SPE, predicted upper stratospheric increases in NO_y over 100% during and shortly after the October 1989 SPE cause significant upper stratospheric O_3 decreases ($> 10\%$). Fluxes of protons used in the computations of NO_y production for the October 1989 events had energies up to 440 MeV [see V96]. Thus energy deposition of these particles and, subsequently NO_y production, occurred down to 50 mbar.

The downward motion in the late fall and winter caused the very large enhancements of NO_y in the upper stratosphere to be transported to lower stratospheric levels, with corresponding greater ozone decreases. Ozone decreases down to 100 mbar were predicted by the spring of 1990 as a result of this event in October 1989. Our predicted

O₃ decrease of over 10% in the upper stratosphere in December 1989 is similar to the NOAA 11 Solar Backscatter Ultraviolet 2 (SBUV/2) measurements of depletions of 12% in the latitude band 60°-80°N at 4 mbar reported in *Jackman et al.* [1995].

Our model transport is climatologically based and probably does not represent the exact transport for the 1989-90 time period [see further discussion of this in *Jackman et al.*, 1995]. The model, however, does a very credible job of simulating several atmospheric tracers [*J96; Fleming et al.*, 1999]. We, therefore, expect that the long-term transport over the 1989-92 period to be reasonable.

Like the August 1972 SPE, the influence of the October 1989 SPE carries on for years past the event. Enhancements of NO_y greater than 5% are predicted to last for two and a half years past the events. Separate SPEs in 1991 that influence the upper stratosphere also do impact the middle stratospheric NO_y and O₃ abundances. Most of the lower stratospheric influence in the 1989-92 time period is a result of the October 1989 SPE. The lower part of the stratosphere (below about 40 mbar) shows enhancements in ozone, especially in the spring and summer period.

Generally, the change in ozone with respect to time at a certain grid point in the model can be written

$$dO_3/dt = \text{Net(Chemistry)} + \text{Net(Transport)}$$

where dO_3/dt is the change in ozone (in mixing ratio / sec), Net(Transport) is the ozone forcing from transport, and Net(Chemistry) is the ozone forcing from chemistry. We set

$$\text{Net(Chemistry)} = \text{Net(P+L)}$$

where “P” is the computed chemical production of ozone and “L” is the computed chemical loss of ozone. Values for “P” are positive and values for “L” are negative.

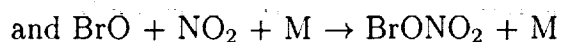
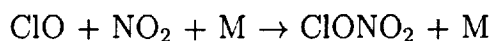
Thus, the change in ozone with respect to time at a model grid point is

$$dO_3/dt = \text{Net}(P+L) + \text{Net}(\text{Transport})$$

We use these definitions for our modelled ozone grid changes to simplify the discussion of our comparison of the “Base” and “Perturbed” simulations.

Results of a comparison between our model simulations are given for 70 mbar and 75°N in Figure 7. Ozone and changes in ozone production and loss rates are shown here over the 1989-92 time period. All the plots in this figure indicate values of ozone production and loss rate changes between the “Base” and the “Perturbed” simulation. The top plot indicates the changes in the two ozone loss rates, $\Delta L(\text{Cl}_x \text{ \& } \text{Br}_x)$ and $\Delta L(\text{NO}_x)$, which are impacted by the SPEs. Changes in other ozone loss rates at this level, such as $\Delta L(\text{HO}_x)$ and $\Delta L(\text{O}_x)$, are not impacted significantly by the SPEs. A positive change indicates a decrease and a negative change indicates an increase in the relevant ozone loss rate.

The loss rate due to the halogens, $L(\text{Cl}_x \text{ \& } \text{Br}_x)$, is changed when the downward transported NO_y interferes with the chlorine and bromine radicals through the reactions



producing more reservoir constituents at the expense of the halogen radicals. $\Delta L(\text{Cl}_x \text{ \& } \text{Br}_x)$ is computed from the relation

$$\Delta L(\text{Cl}_x \& \text{Br}_x) = L(\text{Cl}_x \& \text{Br}_x)_{\text{Perturbed}} - L(\text{Cl}_x \& \text{Br}_x)_{\text{Base}}$$

The $L(\text{NO}_x)$ is changed when the downward transported NO_y leads to a direct increase in the ozone loss from the NO_x constituents. $\Delta L(\text{NO}_x)$ is computed from the relation

$$\Delta L(\text{NO}_x) = \Delta L(\text{NO}_x)_{\text{Perturbed}} - \Delta L(\text{NO}_x)_{\text{Base}}$$

The second and third plots show the change in the total ozone loss and production rates, $\Delta L(\text{total})$ and $\Delta P(\text{total})$, respectively. The fourth plot indicates the net chemical ozone forcing, $\Delta \text{Net}(P+L)$, which is primarily driven by $\Delta L(\text{total})$ changes.

The bottom plot in Figure 7 illustrates the percentage change in ozone (solid line) caused by the SPEs. The forcings for ozone which drive these changes are from both transport and chemistry. The change in ozone with respect to time at a certain grid point in the model can be written

$$d\Delta \text{O}_3/dt = \Delta \text{Net}(P+L) + \Delta \text{Net}(\text{Transport})$$

where $d\Delta \text{O}_3/dt$ is the change in ozone (in mixing ratio / sec), $\Delta \text{Net}(\text{Transport})$ is the ozone forcing from transport, and $\Delta \text{Net}(P+L)$ is the ozone forcing from chemistry. We normalize the change in transport and chemistry terms by dividing through by the background ozone levels and rewrite in units of percentage change per month. These transport and chemistry terms are shown in the bottom plot of Figure 7 and they are represented by the light dotted and long dashed lines, respectively. The transport term is most important in the winter and early spring time period when prevailing downward winds bring air that is somewhat depleted in ozone, forcing a negative change in ozone

at 70 mbar. The chemistry term depends on the sun to drive the photochemical reactions that affect ozone. Changes in the halogen loss processes for ozone, $\Delta L(\text{Cl}_x \& \text{Br}_x)$, occur primarily in the spring, whereas changes in the odd nitrogen loss processes for ozone, $\Delta L(\text{NO}_x)$, occur primarily in the summer.

Simulations over the 1965-95 Time Period

After analyzing the two time periods, 1972-75 and 1989-92, in which the gigantic SPEs of August 1972 and October 1989 dominated, we now turn to the predicted change in constituents over a larger time period, 1965-95. The predicted changes resulting from SPEs at 75°N are given in Figure 8. The behavior of column ozone, represented in the center plot, is a result of all the changes of ozone at various levels above the ground. We pick two levels, 2 and 70 mbar, and indicate the O_3 change and primary chemical mechanisms that mostly explain the predicted long-term behavior of ozone.

During and for a few months after the SPEs, especially the two huge events in 1972 and 1989, the NO_y is enhanced considerably ($> 100\%$) and the O_3 depleted significantly at 2 mbar ($> 20\%$). This behavior is shown in the top two plots of Figure 8. The predicted depletion of O_3 is not linearly anti-correlated to the predicted enhancement of NO_y . The time of year (mid summer in 1972 and mid fall in 1989) is different for the timing of the gigantic SPEs in 1972 and 1989 and the level of stratospheric chlorine is also quite different (see light dashed line in bottom plot of Figure 8).

The O_3 at 70 mbar also varies during this time period as a result of the SPEs (second from bottom plot in Figure 8). The predicted increases in O_3 at this level are

primarily driven by the change in the ozone loss from the halogen constituents, $\Delta L(\text{Cl}_x \& \text{Br}_x)$, shown in the bottom plot in Figure 8. This term becomes larger as the amount of Cl_y increases in the stratosphere (represented by the light dashed line in the bottom plot in Figure 8). The August 1972 SPE occurred when Cl_y levels were about 1.2 ppbv and the October 1989 SPE occurred when Cl_y levels were nearly 3 ppbv, an increase of a factor of about 2.5.

A positive change in $\Delta L(\text{Cl}_x \& \text{Br}_x)$, shown in the bottom plot of Figure 8, means that the halogen chemical loss for ozone has been decreased. This decrease in halogen chemical loss will force an ozone production, however, this forcing may not actually lead to a net ozone increase at this level. In some years the transport of depleted ozone to lower levels as well as the increase in $\Delta L(\text{NO}_x)$ will overwhelm this decrease in $L(\text{Cl}_x \& \text{Br}_x)$. Ozone increases at 70 mbar are most apparent when Cl_y levels are the largest after the October 1989 SPE (see years 1990-94).

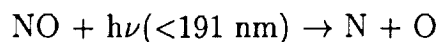
How would the stratosphere have responded to the August 1972 SPE in a high chlorine environment and, conversely, how would the stratosphere have responded to the October 1989 SPE in a low chlorine environment? We attempted to answer these questions with several sensitivity studies which varied the levels of halogen (both chlorine and bromine) and the SAD amounts in the stratosphere. We do not show the results of all the sensitivity studies here, but do show the two extreme cases examined.

Each of these extreme cases included a base and a perturbed simulation and are presented in Table 2. The “Base_{Lo}” simulation was similar to the “Base” simulation, but held chlorine and bromine fixed at low levels and stratospheric SAD fixed at background

levels. The “Base_{Hi}” simulation was similar to the “Base” simulation, but held chlorine and bromine fixed at high levels (corresponding to 1995) and stratospheric SAD fixed at the very high levels measured in 1991-2 after the Mt. Pinatubo eruption. The “Base_{Hi}” stratospheric SAD changed monthly, but repeated yearly simply by recycling the June 1991 through May 1992 measured values. The “Perturbed_{Lo}” and “Perturbed_{Hi}” simulations were the same as “Base_{Lo}” and “Base_{Hi}”, respectively, except they also included solar proton events.

The polar column ozone predicted changes are given in Figures 9a and 9b for the 50°-90°N and 50°-90°S regions, respectively. The solid lines indicate the predicted ozone changes for the primary simulations, “Base” and “Perturbed.” Total ozone decreases greater than 0.5% are computed for the northern polar region in 1990 and for the southern polar region in 1973 and 1990. Larger effects are computed in the southern hemisphere in 1973 and in the northern hemisphere in 1990. The different hemispheric influences are caused by the different months of occurrences for the gigantic SPEs in 1972 and 1989.

The August 1972 SPE occurred in the middle of winter in the southern hemisphere. At that time of year the much lower sunlight in the polar region conserves the SPE-generated NO_y since the primary upper and middle stratospheric NO_y loss mechanism is



followed by $\text{N} + \text{NO} \rightarrow \text{N}_2 + \text{O}$

This allows more of the NO_y to be transported by the predominant downward moving winds to low levels. The higher sunlight and upward motion in the northern polar summer would lead to very significant NO_y destruction. Thus the southern polar region shows a larger ozone impact than the northern polar region during this event.

The October 1989 SPE occurred in the middle of fall in the northern hemisphere and the middle of spring in the southern hemisphere. The northern hemisphere at this time is experiencing a decrease in sunlight, accompanied by a decrease in the NO_y destruction rate, and a slow change to a prevailing downward motion. In contrast, the southern hemisphere is experiencing an increase in sunlight, accompanied by an increase in the NO_y destruction rate, and a slow change to a prevailing upward motion. These interhemispheric differences lead to a slightly larger ozone decrease predicted in the northern hemisphere.

The dashed lines in Figure 9 represent the predicted ozone changes for the simulations, "Base_{Lo}" and "Perturbed_{Lo}." These simulations, with the lowest levels of halogens and smallest background SAD, predict the largest ozone depletions. The combination of these two changes leads to an enhanced importance of NO_x constituents in control of ozone and significantly more predicted ozone depletion, especially after the October 1989 SPE. The NO_x radicals NO and NO_2 are enhanced in the decreased halogen environment due to a decrease in the ClONO_2 and BrONO_2 reservoirs and, therefore, an increase in the NO_x radicals. In 1990 the northern hemisphere shows ozone depletion increasing from -0.9 to -1.4% and the southern hemisphere shows depletions

increasing from -0.7 to -1.2%, when comparing the “Perturbed” to the “Base” simulation and the “Perturbed_{Lo}” to the “Base_{Lo}” simulation, respectively.

The dotted lines in Figure 9 represent the predicted ozone changes for the simulations, “Base_{Hi}” and “Perturbed_{Hi}.” These simulations, with the highest levels of halogens and largest background SAD, predict the smallest ozone depletions or ozone enhancements. The combination of these two changes leads to an enhanced importance of the halogen constituents in control of ozone and significantly less predicted ozone depletion, especially after the August 1972 SPE. In 1973 the northern hemisphere shows ozone depletion reduced from -0.35 to -0.2% and the southern hemisphere shows depletions reduced from -0.8 to -0.25%, when comparing the “Perturbed” to the “Base” simulation and the “Perturbed_{Hi}” to the “Base_{Hi}” simulation, respectively. Enhancements in ozone are predicted for years 1974-76 after the August 1972 SPE in these simulations and for years 1991-94 after the October 1989 SPE.

Another important atmospheric change, which will have a significant impact on the influence of huge SPEs on the stratosphere, is the interannual variability of the transport. The very consequential role that transport has in determining the influence of SPEs on the stratosphere was discussed in *J95*. A winter with substantially larger polar downwelling shortly after a gigantic SPE (such as the northern winter of 1989-90) will convey the SPE-produced NO_y constituents to lower stratospheric levels. At these lower stratospheric levels the SPE-produced NO_y constituents will be more effectively protected from sunlight and thus able to influence the stratosphere over a longer period of time. We did not investigate this significant transport influence on the SPE effects,

as it is outside the scope of this study.

Conclusions

The extremely large particle events in August 1972 and October 1989 occurred in very chemically different stratospheres. The August 1972 SPE impacted the stratosphere when chlorine levels were much lower than those of the stratosphere in October 1989. These huge events affected total ozone for several months to years past the events by causing direct ozone decreases from NO_y -induced losses. Because of the seasonal variations in transport and solar illumination, polar ozone decreases are predicted to be larger for the gigantic SPEs occurring in the fall or winter, and smaller decreases for SPE events occurring in the spring or summer. Interference of the NO_y constituents with the halogen loss cycles for ozone destruction actually led to some total ozone production that was especially notable in the time period 1992-4, a few years after the October 1989 SPE.

If the August 1972 SPE had occurred in a more halogen-controlled stratosphere, the predicted total ozone decreases from this SPE would have been diminished with ozone increases predicted several years after the event. Conversely, if the October 1989 SPE had occurred in a stratosphere with less halogen constituents, the predicted total ozone decreases from this SPE would have been enhanced.

Since the halogen amounts in the stratosphere are expected to decrease only very slowly over the next several decades from their present-day levels, the simulations of the October 1989 SPE are believed to be most relevant for future extremely large

SPEs in the next couple of solar cycles. These future events will probably initially decrease ozone substantially in the upper stratosphere and then gradually impact the lower stratospheric ozone in a minor way several months to years past the events by interference with the halogen loss cycles.

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Figure Captions

Figure 1. Total number of NO_y molecules produced per year in the northern polar stratosphere by SPEs (histogram indicating both the total and the stratospheric contribution), GCRs (dash-dot-dot-dot line), N_2O oxidation in the polar region only (dash-dot line), and horizontal transport of NO_y from lower latitudes into this region (dashed line).

Figure 2. Model computed percentage total ozone changes from 1972-1975 resulting from SPEs. Contour intervals are .0, -.1, -.2, -.5, -.8, -1., -1.2, and -2.%

Figure 3. Model computed percentage total ozone changes from 1989-1992 resulting from SPEs. Contour intervals are +.2, +.1, .0, -.1, -.2, -.5, -.8, -1., -1.2, and -2.%

Figure 4. Model computed percentage changes in NO_y and O_3 at 75°N for 1972-1975 resulting from SPEs. Contour intervals for NO_y are 0, 1, 3, 5, 10, 30, 100, and 200%. Contour intervals for O_3 are +3., +2., +1., +.5, +.2, .0, -.2, -.5, -1., -2., -5., -10., -20.%

Figure 5. Model computed percentage changes in NO_y at 2 mbar, O_3 at 2, 12, and 30 mbar, and column O_3 for 75°N in 1972 resulting from SPEs. The abscissa is the same, however, the ordinate is different in each plot.

Figure 6. Model computed percentage changes in NO_y and O_3 at 75°N for 1989-1992 resulting from SPEs. Contour intervals for NO_y are 0, 1, 3, 5, 10, 30, 100, and 200%. Contour intervals for O_3 are +.5, +.2, .0, -.2, -.5, -1., -2., -5., -10., -20.%

Figure 7. Model computed changes in ozone rates (in mixing ratio $\times 10^{16}$ / sec) resulting from SPEs is represented in the top four plots at 70 mbar and 75°N . Model computed percentage change in O_3 (solid line) and percentage change per month in Net(Transport) and Net(P+L) at 70 mbar and 75°N resulting from SPEs is represented by the light dotted and light dashed lines, respectively.

Figure 8. Model computed percentage changes in NO_y and O_3 at 2 mbar, column O_3 , O_3 at 70 mbar, and the change in $L((\text{Cl}_x \text{ \& } \text{Br}_x))$ at 70 mbar resulting from SPEs for 75°N are represented by the solid lines in the five plots for the time period 1965-95. The units for the "Change in Rate" given in the bottom plot are mixing ratio $\times 10^{16}$ / sec. The bottom plot also has the global average Cl_y level at 1 mbar represented by the light dashed line and the values given on the right ordinate.

Figure 9. The total ozone percentage change predicted to be caused by the SPEs in the (a) northern polar region and (b) southern polar region over the 1965-95 time period. The solid, dashed, and dotted lines are a result of comparing simulations "Perturbed" to "Base", "Perturbed_{Lo}" to "Base_{Lo}", and "Perturbed_{Hi}" to "Base_{Hi}", respectively.

Table 1. Description of primary time dependent model simulations

Simulation	Description
Base	No solar proton events included; Chlorine and bromine source gases from <i>WMO</i> [1999]; Stratospheric SAD from <i>J96</i>
Perturbed	Same as “Base” except solar proton events included

Table 2. Description of other time dependent model simulations

Simulation	Description
Base _{Lo}	<p>No solar proton events included;</p> <p>Chlorine only from CH₃Cl fixed at 550 pptv;</p> <p>Bromine only from CH₃Br fixed at 9 pptv;</p> <p>Stratospheric SAD at <i>WMO</i> [1992] background levels</p>
Perturbed _{Lo}	Same as “Base _{Lo} ” except solar proton events included
Base _{Hi}	<p>No solar proton events included;</p> <p>Chlorine source gases fixed at 1995 levels from <i>WMO</i> [1999];</p> <p>Bromine source gases fixed at 1995 levels from <i>WMO</i> [1999];</p> <p>Stratospheric SAD using high 1991-2 levels</p>
Perturbed _{Hi}	Same as “Base _{Hi} ” except solar proton events included

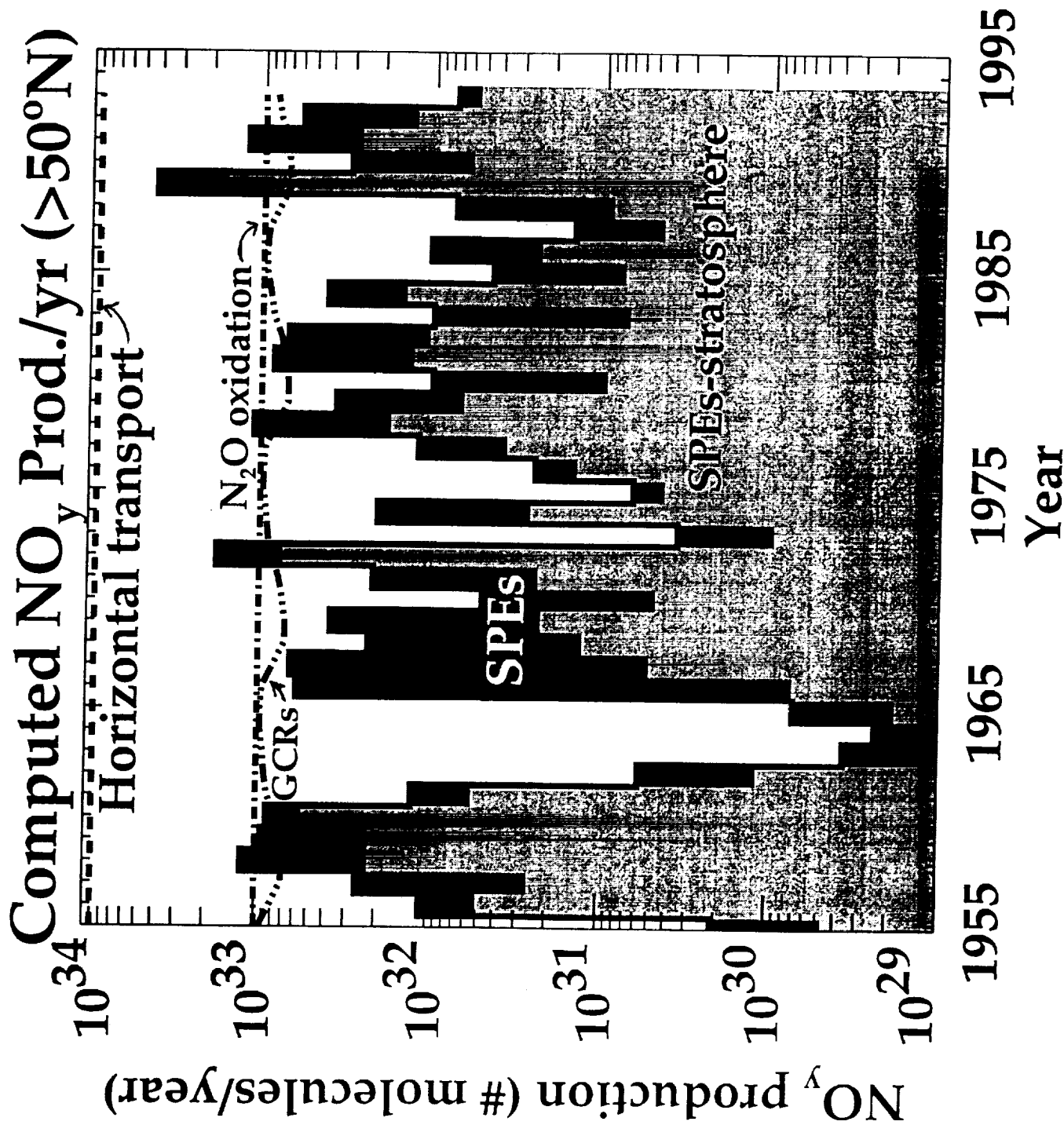


Figure 1. Total number of NO_y molecules produced per year in the northern polar stratosphere by SPES (histogram indicating both the total and the stratospheric contribution), GCRs (dash-dot-dot line), N_2O oxidation in the polar region only (dash-dot line), and horizontal transport of NO_y from lower latitudes into this region (dashed line)

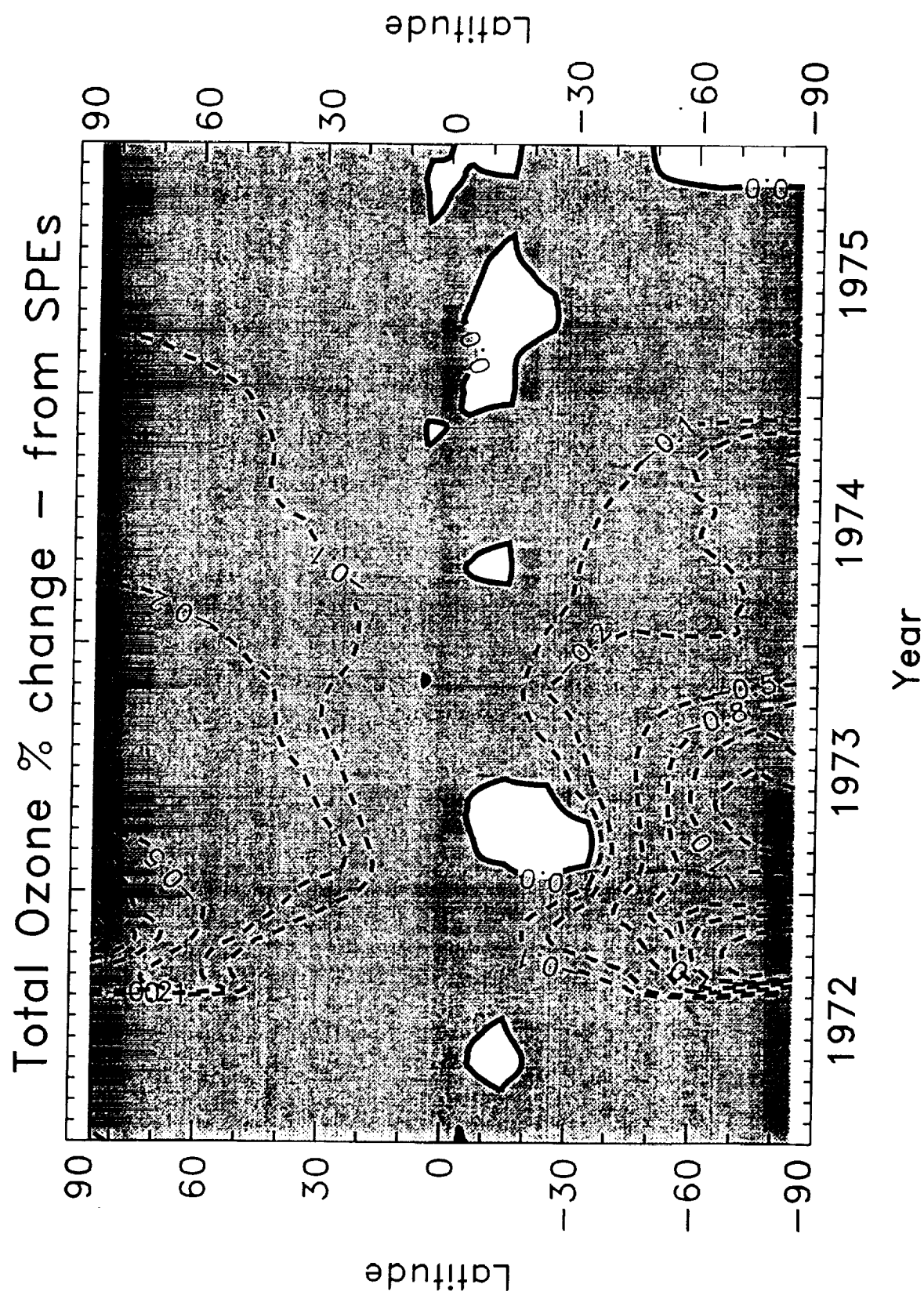


Figure 2. Model computed percentage total ozone changes from 1972-1975 resulting from SPEs. Contour intervals are .0, -.1, -.2, -.5, -.8, -1., -1.2, and -2.%

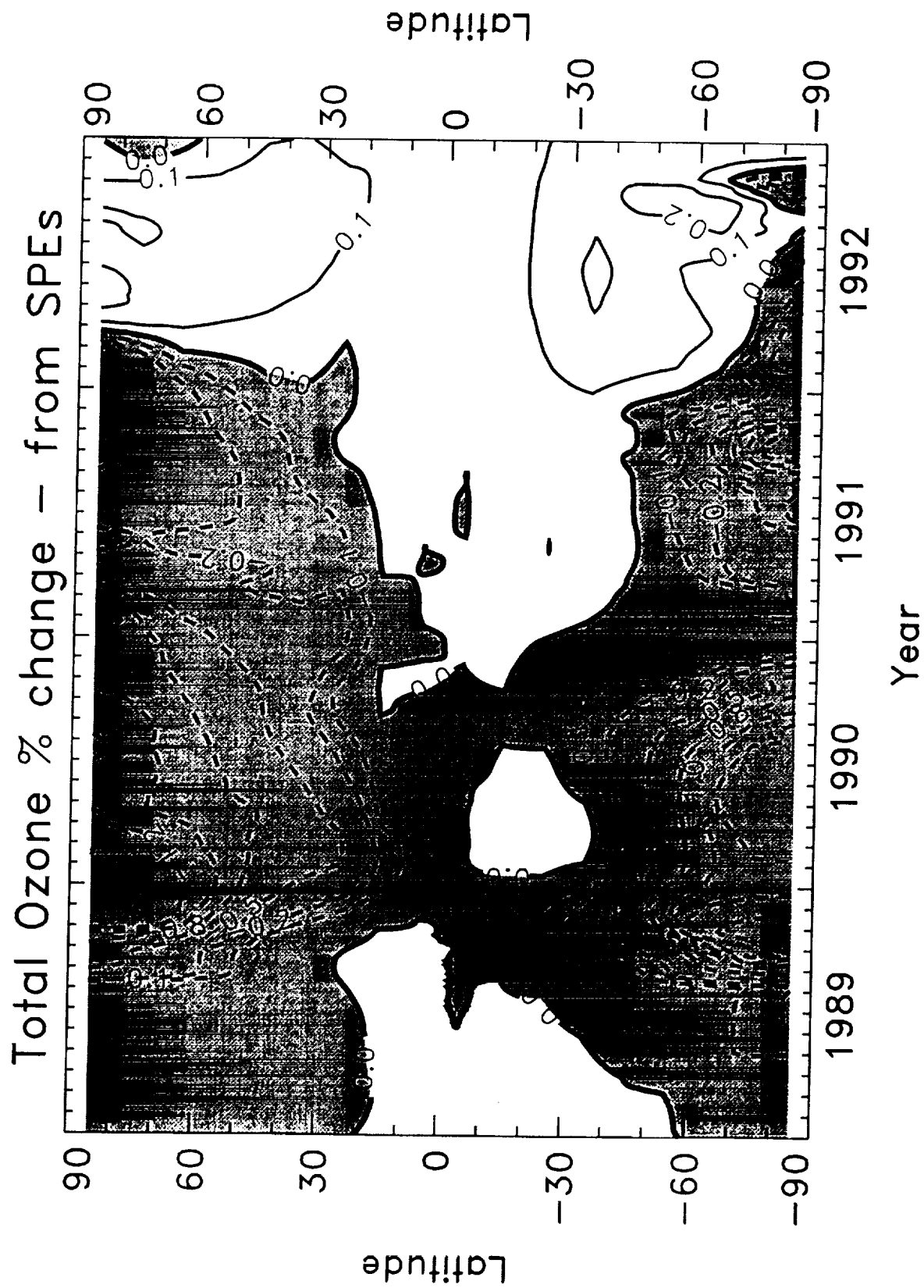


Figure 3. Model computed percentage total ozone changes from 1989-1992 resulting from SPEs. Contour intervals are +.2, +.1, .0, -.1, -.2, -.5, -.8, -1., -1.2, and -2. %

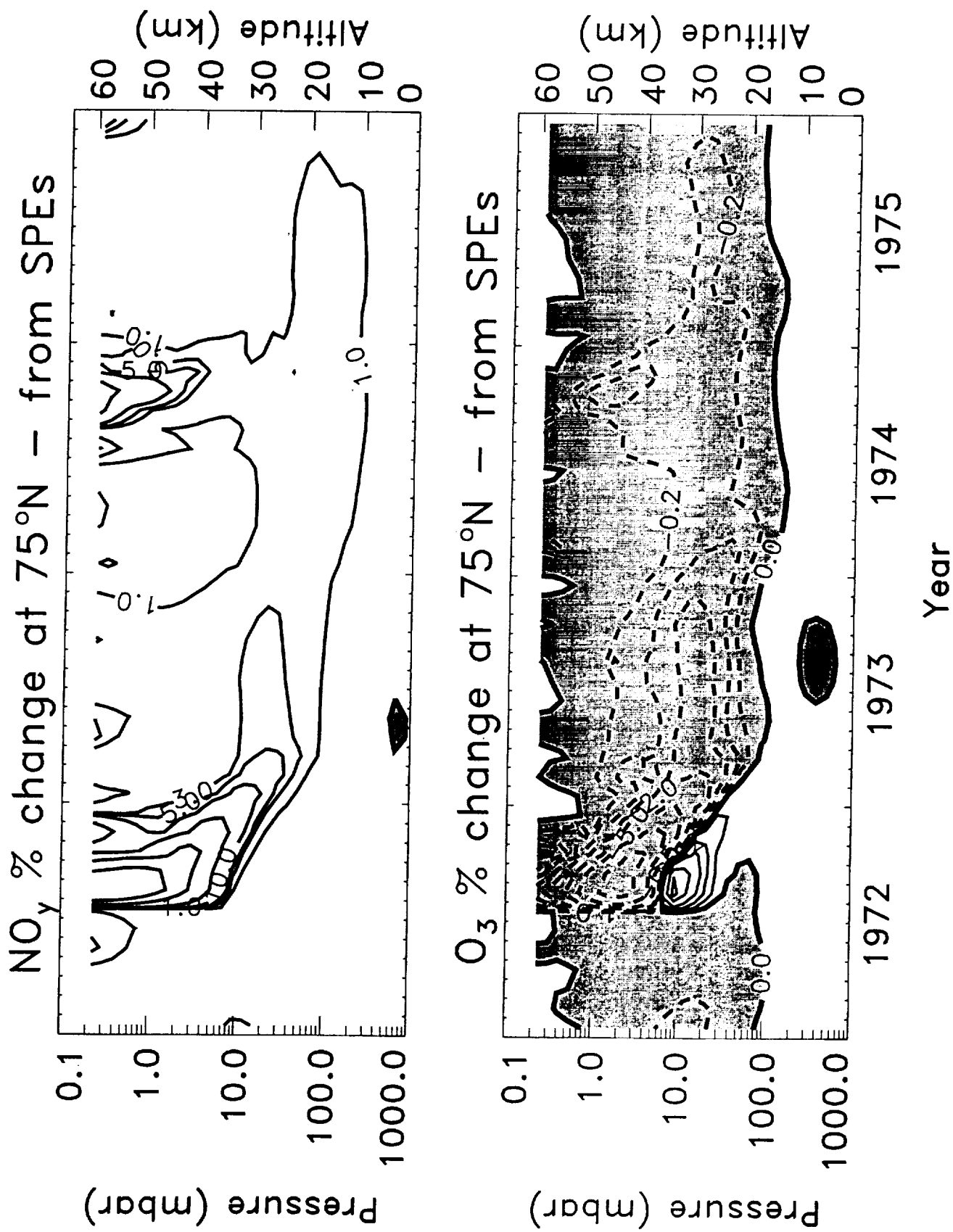


Figure 4. Model computed percentage changes in NO_y and O₃ at 75°N for 1972-1975 resulting from SPEs. Contour intervals for NO_y are 0, 1, 3, 5, 10, 30, 100, and 200%. Contour intervals for O₃ are +3., +2., +1., +.5, +.2, .0, -.2, -.5, -1., -2., -5., -10., -20.%

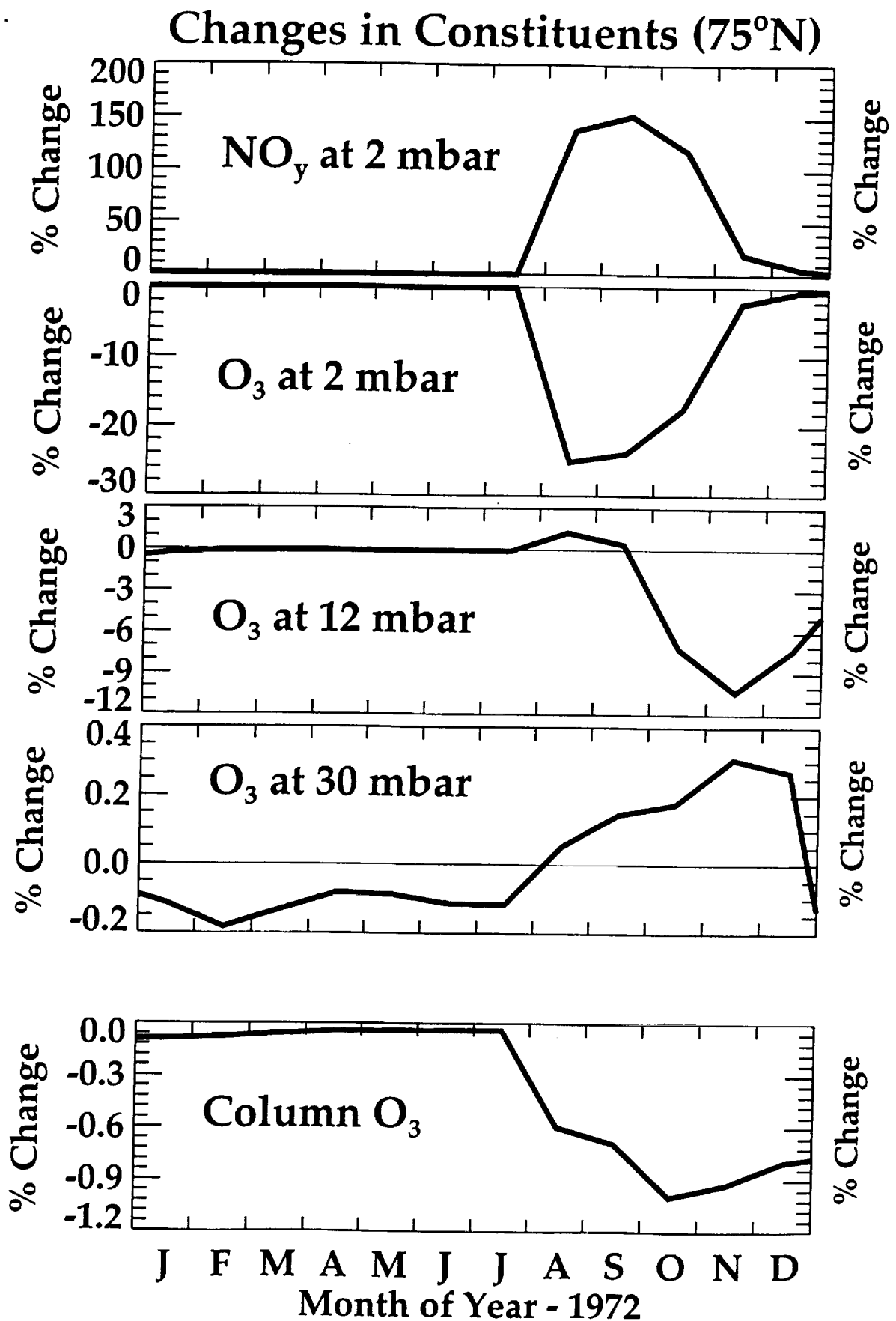


Figure 5. Model computed percentage changes in NO_y at 2 mbar, O₃ at 2, 12, and 30 mbar, and column O₃ for 75°N in 1972 resulting from SPEs. The abscissa is the same, however, the ordinate is different in each plot.

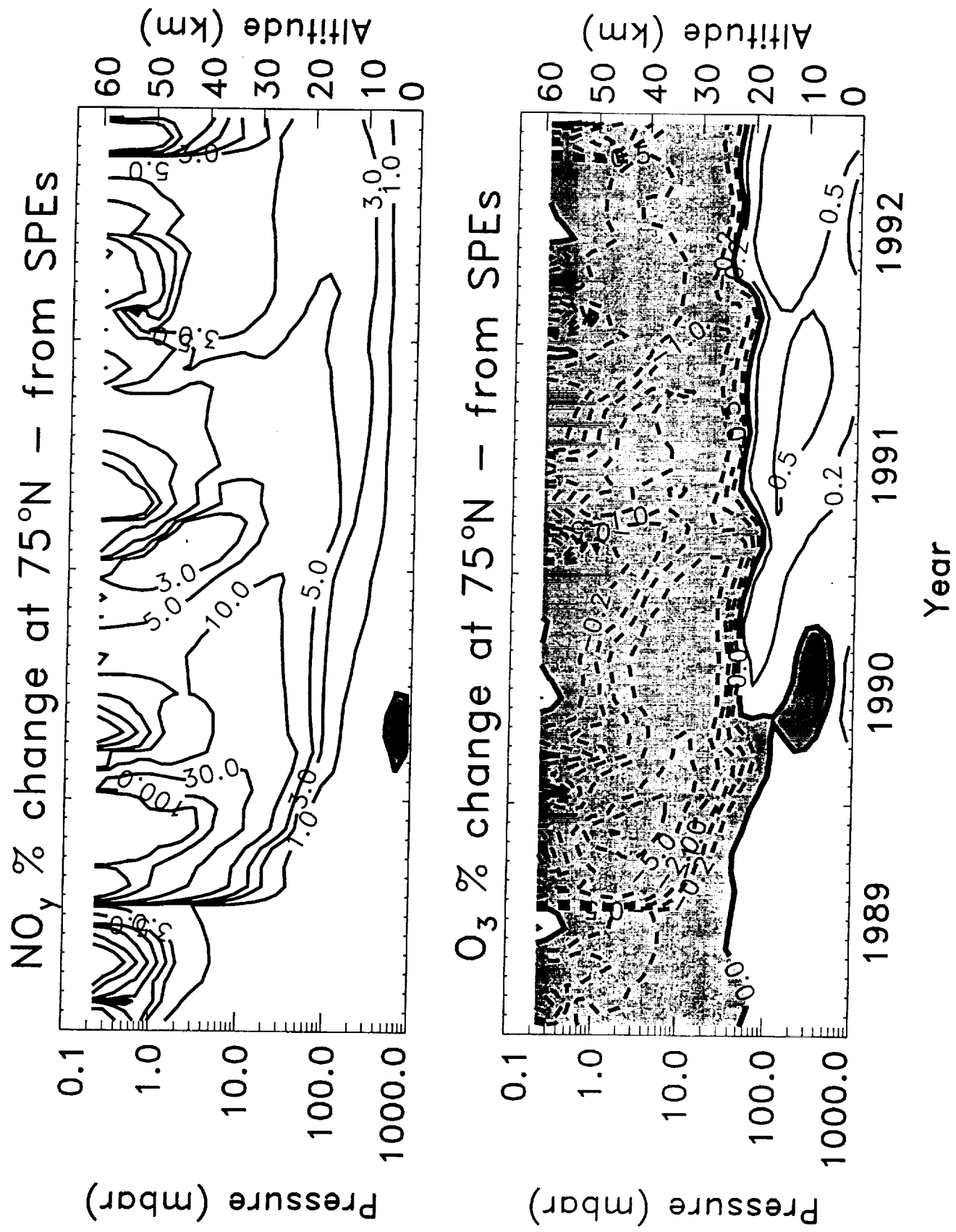
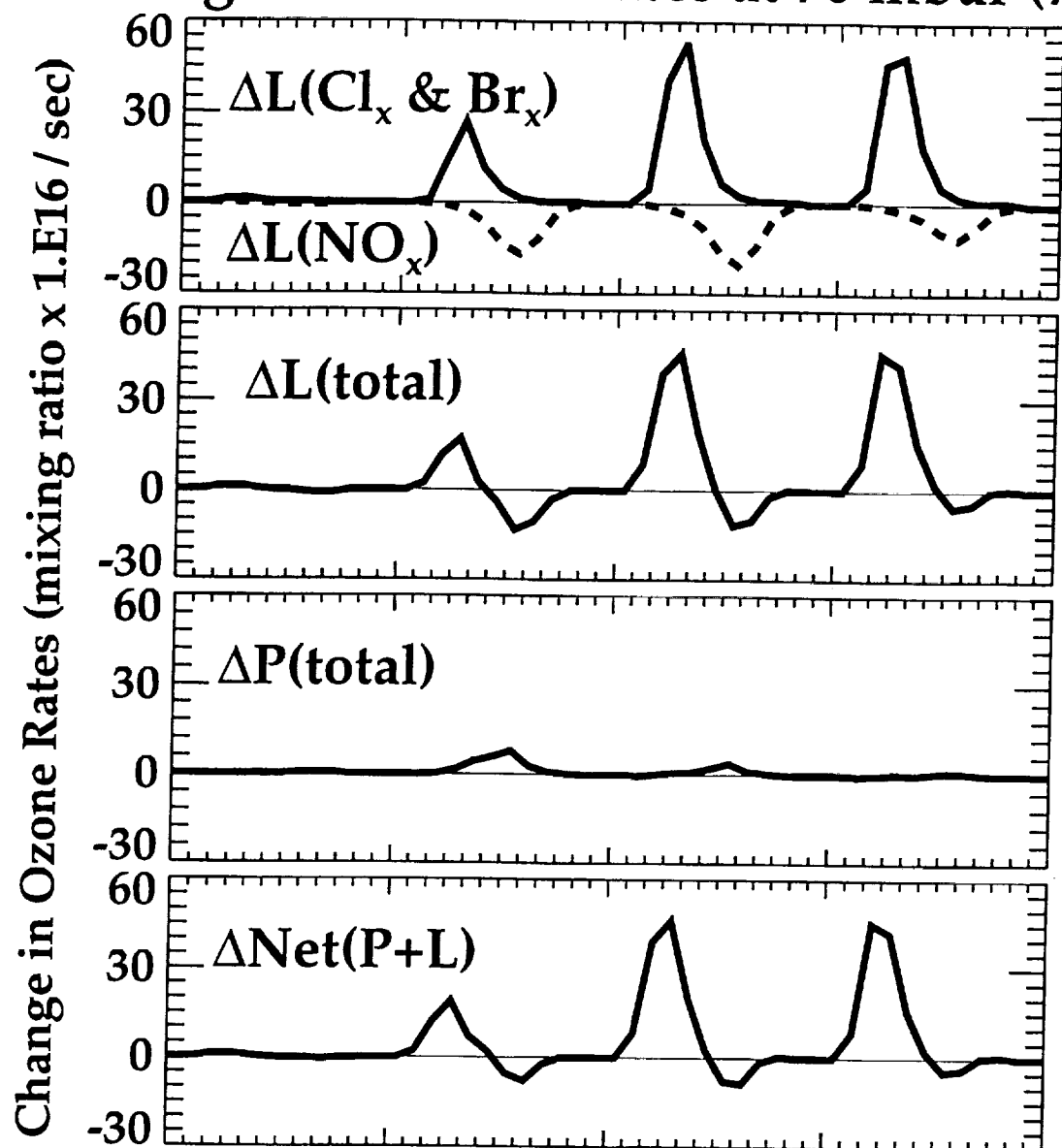


Figure 6. Model computed percentage changes in NO_y and O₃ at 75°N for 1989-1992 resulting from SPEs. Contour intervals for NO_y are 0, 1, 3, 5, 10, 30, 100, and 200%. Contour intervals for O₃ are +5, +2, 0, -2, -5, -1, -2, -5, -10, -20, -50, -100, and -200%.

Changes in Ozone Rates at 70 mbar (75°N)



Change in O_3 at 70 mbar (75°N)

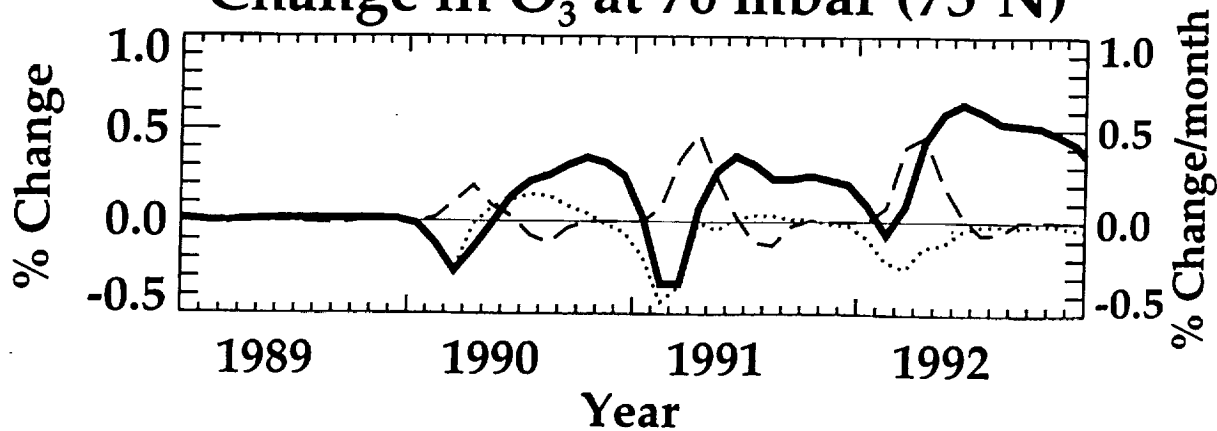


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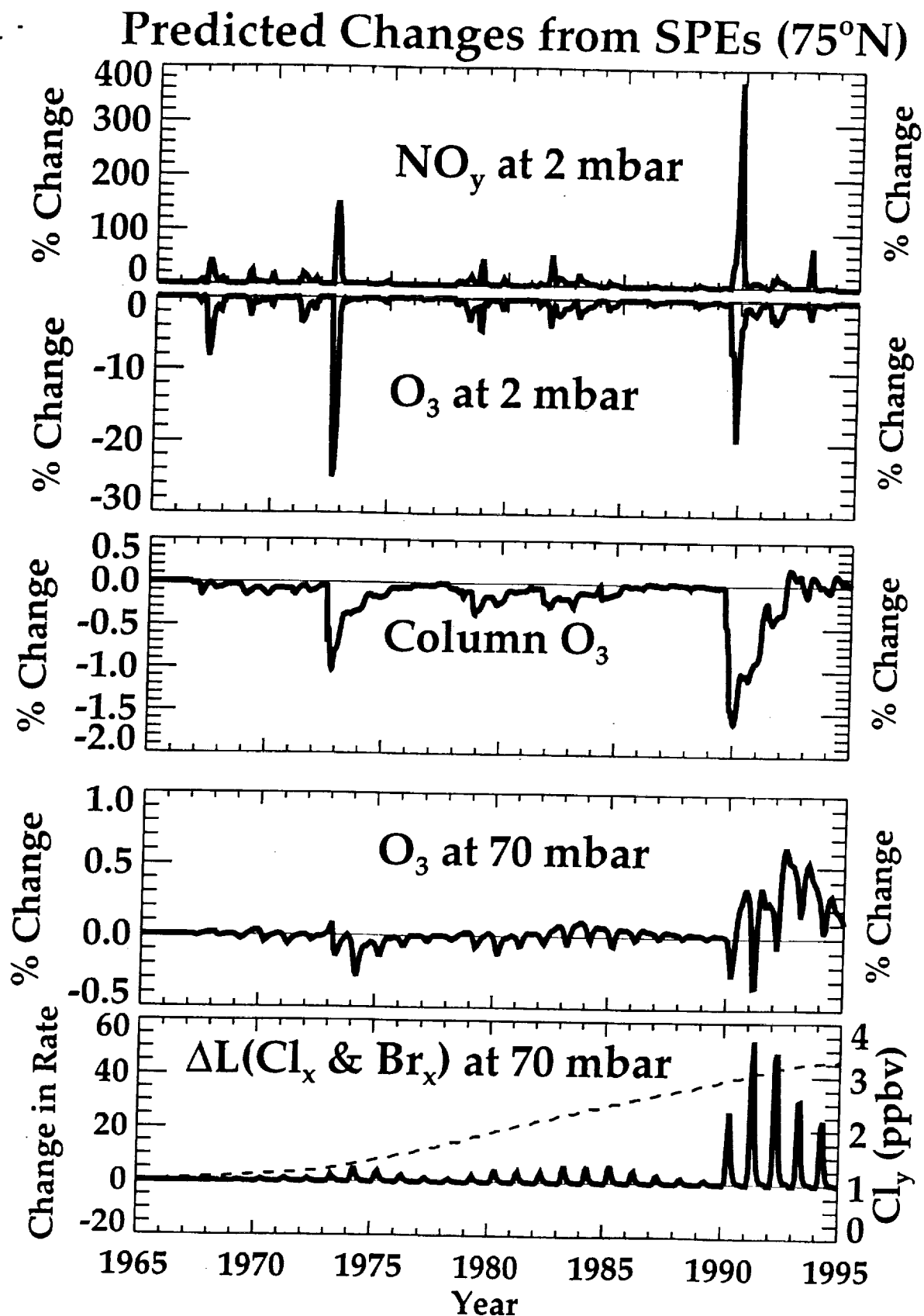
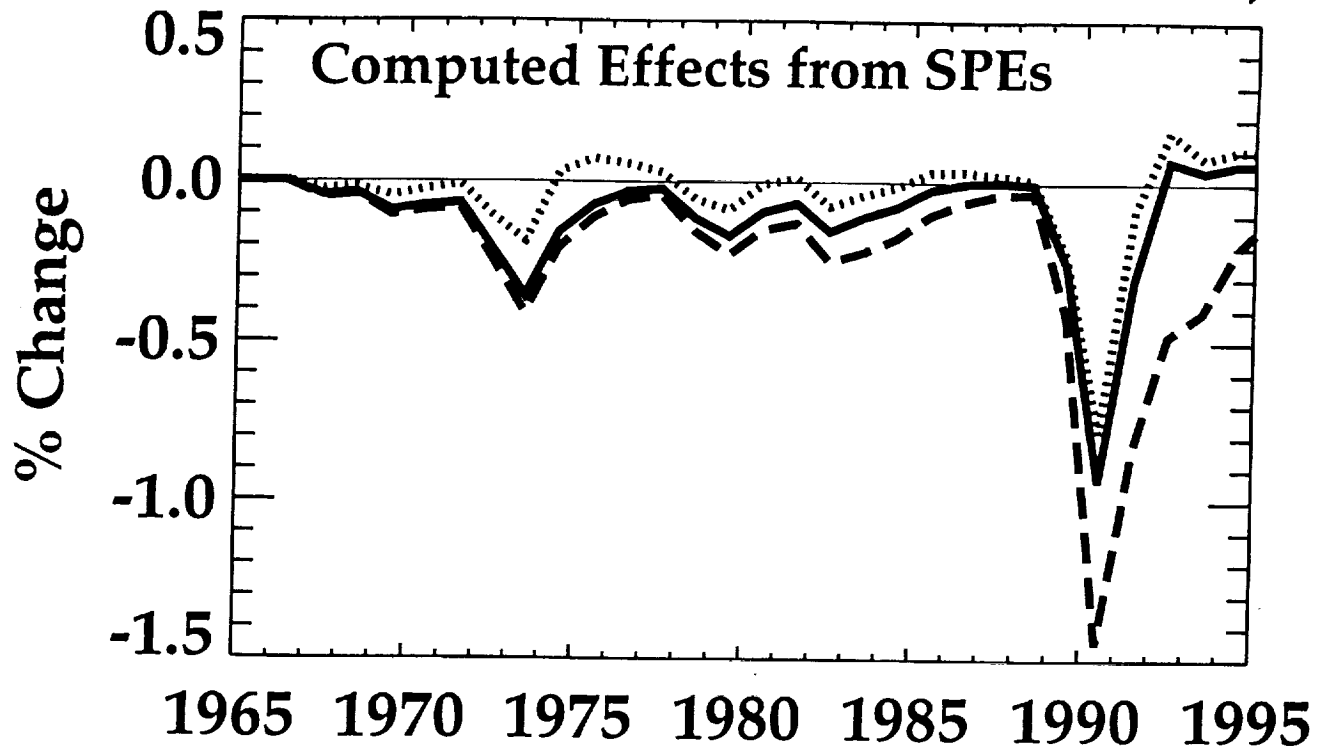


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(a) Total Ozone (50° to 90° N)



(b) Total Ozone (50° to 90° S)

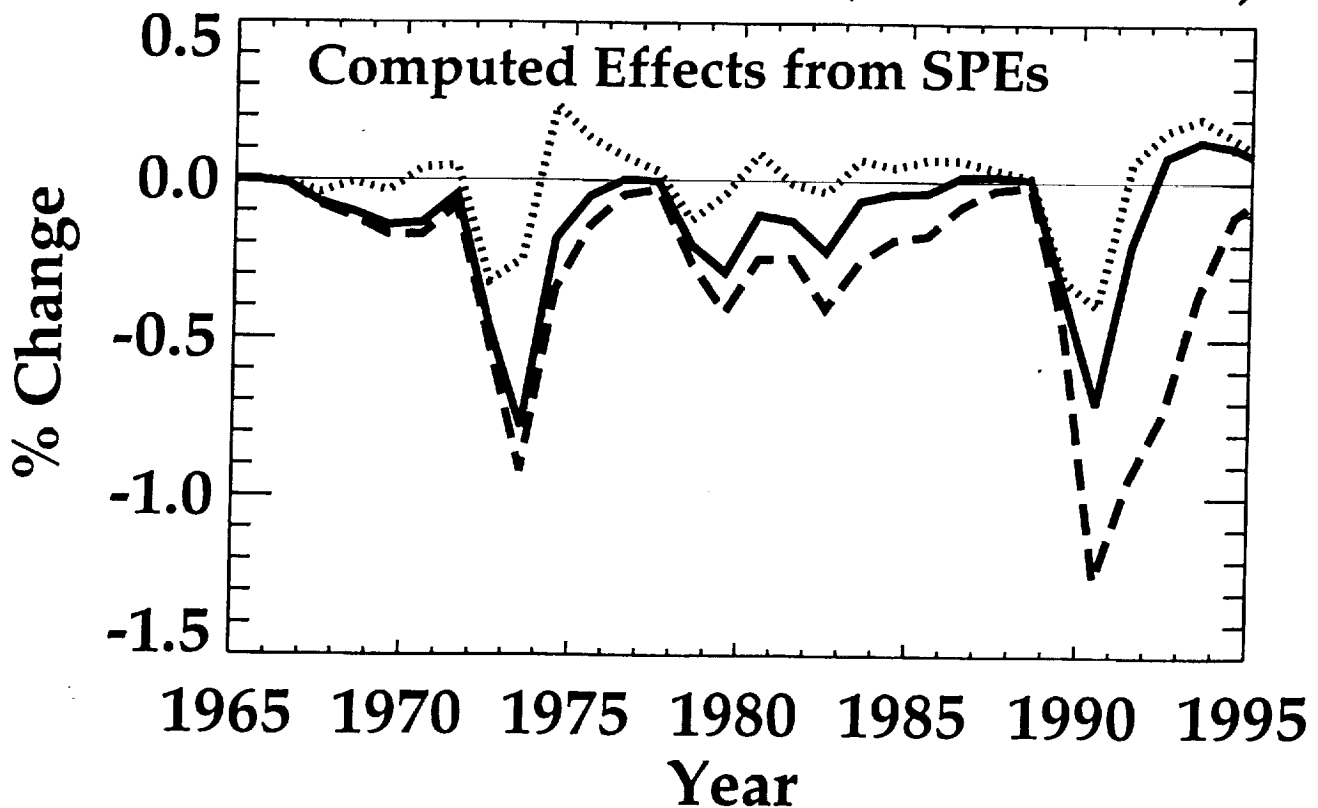


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